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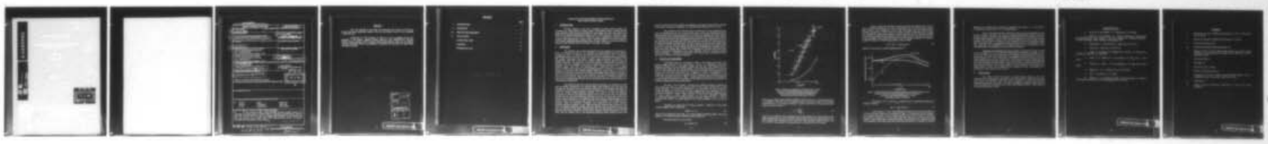

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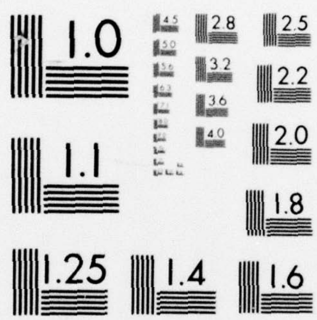

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(U) We have measured, at the 10-μm micrometer wavelength, anomalous infrared emission from condensing and cooling clouds of saturated steam at atmospheric pressure.* Radiant emittance values were observed which were three to four times those calculated for water droplets and water vapor present in the clouds. Near 100°C, luminescence-like activity was observed confirming earlier observations by other workers.

* was measured.

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PREFACE

The work described in this report was authorized under Project 1T161101A91A, In-house Laboratory Independent Research. This work was started in June 1976 and completed in September 1977.

CONTENTS

ANOMALOUS INFRARED EMISSION FROM CONDENSING AND COOLING STEAM CLOUDS

I. INTRODUCTION.

We have measured, at the 10-μm wavelength, anomalous infrared emission from condensing and cooling clouds of saturated steam at atmospheric pressure. Radiant emittance values were three to four times those calculated for water droplets and water vapor present in the clouds over the experimental temperature range $\theta_C = 30°$ to $100°C$ and droplet concentration range $C = 0.1$ to $3.0\ gm/m^3$. At 100°C and $C = 3.0\ gm/m^3$ under nonequilibrium conditions, radiant emittance values approximately 1.4 times those of blackbody radiators at the same temperature were recorded, suggesting luminescence-like activity of water. Evidence for such activity has been reported by other workers both in the infrared[1,2] and visible[3] wavelengths.

II. PROCEDURE.

The equipment used in these measurements has been described by us previously.[4,5] Steam was introduced into the top of a steel-walled, externally-insulated, vertical test chamber 3 m high and 3.05 m in diameter. The chamber walls were provided with 15-cm-diameter, diametrically-opposed view ports which were covered with infrared-transmitting polyethylene film windows. Precautions were taken to prevent steam condensation on windows. Steam measurements made with or without the windows therefore produced identical results. An infrared radiometer[4] was placed at one view port opposite a thermostatted, water-jacketed, horizontal steel cone blackbody which had an optical aperture 30 cm in diameter and an emissivity greater than 0.98. The radiometer had a field-of-view (FOV) of one degree (FOV = 17 mrad) and was used at a fixed wavelength setting of $\lambda = 10\ \mu m$ for the measurements described in this paper. The chamber diameter defined the optical path, L, as 3.05 m. A He:Ne ($\lambda = 0.63\ \mu m$) laser and power meter detector were aligned along an optical axis parallel to that of the radiometer and the steel cone blackbody. The laser and radiometer readings thus could be used to determine steam droplet size and concentration.[6] Gravimetric samplers were also used for obtaining droplet diameter and size distribution information using sampling probes located along the optical axes.

The radiometer also had an internal reference blackbody, the temperature of which could be precisely determined by an electronic bridge. Initial calibration runs were made with the radiometer viewing the steel cone blackbody through the 3.05-m-chamber optical path. The temperature of the steel cone blackbody was slowly raised from 25° to 100°C, and calibration curves were obtained relating radiometer response at $\lambda = 10\ \mu m$ to the steel cone blackbody temperature for the steel cone blackbody emissivity of 0.98. For the actual steam measurements, however, the steel cone (target) blackbody temperature was kept exactly the same as that of the internal reference blackbody in the radiometer. This insured that no radiometric signal could be detected except from the steam clouds introduced into the chamber. This "radiometric null" condition, which was maintained throughout each experimental trial, allowed maximum radiometer sensitivity settings to be used. Thermocouples and calibrated glass thermometers were used to determine temperature profiles within the chamber along the optical axes and at the steel cone blackbody water jacket. Thermocouples were also used between the steel chamber walls and their exterior insulation. The time rate of change of chamber temperature was much slower than the response time of the temperature-measuring devices during the steam cloud

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cool-down periods. In the null condition, the radiometer could detect radiant emittance changes corresponding to changes in temperature, θ_c, of the steel cone blackbody of 0.2°C or less.

After radiometric null conditions had been achieved prior to each experimental trial, steam was introduced into the top of the chamber from a valved pipe. Chamber fans insured uniform mixing. Steam introduction lasted from 2 to 30 minutes, depending upon initial chamber wall temperature and the desired temperature range of the steam cloud which would be allowed to cool after steam shutoff. The wall temperature reached 70°C in some experiments but for the data reported here was initially near 50°C. When steam flow was shut off, the temperature of the steam cloud within the chamber fell to 35°–40°C within a few minutes, and the wall cooled to 40°-45°C over the same time period. During the very interesting cool-down periods which lasted 30 to 40 minutes in many experiments, wall and cloud temperatures converged with the last residual steam droplets seen at about 30°C, illuminated by the He:Ne laser beam. Steam droplet concentration determinations were made repeatedly during the cool-down periods. Saturation humidity, i.e., 100% RH, was maintained in the chamber nearly until the end of each trial when the residual water droplets evaporated.

III. RESULTS AND DISCUSSION.

Experimental results are summarized in figure 1. The ordinate is the ratio of the spectral radiant emittance of the cloud, $(W_{10})_\theta$, to that of a blackbody at the same temperature, $(W^o_{10})_\theta$, for the wavelength $\lambda = 10\ \mu m$. Since the steel cone blackbody had an emissivity greater than 0.98, the ratio $(W_{10}/W^o_{10})_\theta$ is very nearly the emissance, ϵ, of the steam *cloud for the experimental data shown* by the upper curve labeled with temperatures and showing error bars. The vertical error bars correspond to errors in measurement of the temperature of the steam cloud in the chamber of ±3°C. Actual error was not believed to exceed ±1.5°C. The horizontal error bars of figure 1 represent maximum errors in the determination of steam droplet concentration C and include the full ranges of error both for the optical[6] and gravimetric methods used in these measurements.

The lower curve of figure 1 represents the combined emissance calculated for water droplets and vapor for various abscissa values of water *droplet* concentration, C, times optical path length, L, the product of which has the "CL" units gm/m^2. The vapor contribution to this calculated curve at $\lambda = 10\ \mu m$ is nearly negligible since this wavelength lies in a well-known atmospheric "window" region, well away from major interatomic absorption bands of water vapor or other gases common to the lower atmosphere.[7] Bignell's "k_1" absorption coefficients[8] for pressure-broadened water vapor absorption near $\lambda = 10\ \mu m$ are extremely small. Clearly, the steam droplets are the major contributor to the ϵ_A curve of figure 1.

Kirchhoff's law states that, for bodies in thermal equilibrium (as in the steam cool-down periods of these experiments),

$$(W/W^o) = A = \epsilon \tag{1}$$

where A is the absorptance of the body, e. g., a water droplet in the present paper – that is, the emissance, ϵ, of a body under these conditions equals its absorptance.

The Beer-Lambert law can be written:

$$T_\lambda = \exp(-\alpha_\lambda\ CL), \tag{2}$$

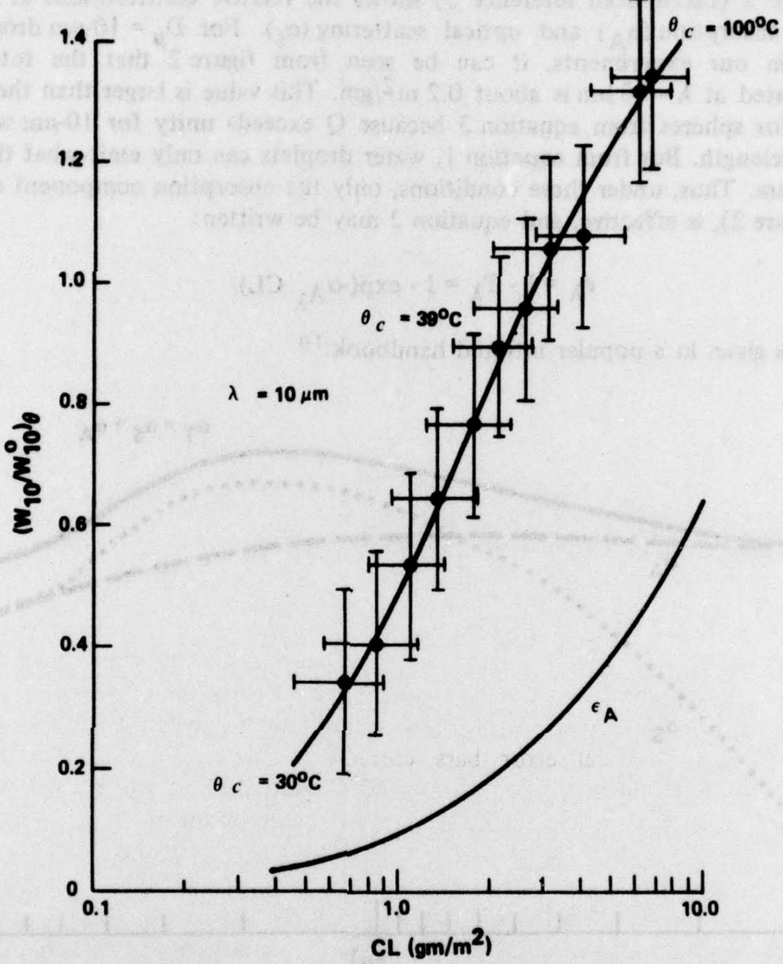

Figure 1. Calculated Curve (Labeled ϵ_A) for Emission by Water Droplets and Vapor at 10-μm Wavelength Compared to Experimental Curve (with Error Bars) for Same Wavelength Showing Magnitude of Anomalous Emission (Optical path length, 3.05 meters)

where α_λ is the droplet mass extinction coefficient, m^2/gm, and T_λ is the optical transmittance at wavelength λ. The value of α for spherical particles is simply the ratio of the geometric cross-sectional area of all droplets contained in a given cloud volume to the mass of material present in the same volume or

$$\alpha = \frac{3\,Q}{2\,D_\mu \rho} \tag{3}$$

where Q is an efficiency factor calculated from the Mie theory,[9] D_μ is the geometric mean droplet diameter, μm, and ρ is the density of the material forming the droplets, gm/cm^3. For water droplets from condensed steam, which have mean geometric droplet diameters near $D_\mu = 10\ \mu m$,[5,6] from equation 3 the expected value of α would be about 0.15 m^2/gm.

Figure 2 (taken from reference 5) shows the relative contributions at $\lambda = 10\ \mu m$ to α_{10} by droplet absorption (α_A) and optical scattering (α_s). For $D_\mu = 10$-μm droplets such as those observed in our experiments, it can be seen from figure 2 that the total extinction coefficient calculated at $\lambda = 10\ \mu m$ is about $0.2\ m^2/gm$. This value is larger than the $0.15\ m^2/gm$ value calculated for spheres from equation 3 because Q exceeds unity for 10-μm water droplets at the 10-μm wavelength. But from equation 1, water droplets can only emit what they absorb in thermal equilibrium. Thus, under these conditions, only the absorption component of extinction, α_A (shown in figure 2), is effective, and equation 2 may be written:

$$\epsilon_\lambda = 1 - T_\lambda = 1 - \exp(-\alpha_{A_\lambda}\ CL). \tag{4}$$

Equation 4 also is given in a popular infrared handbook.[10]

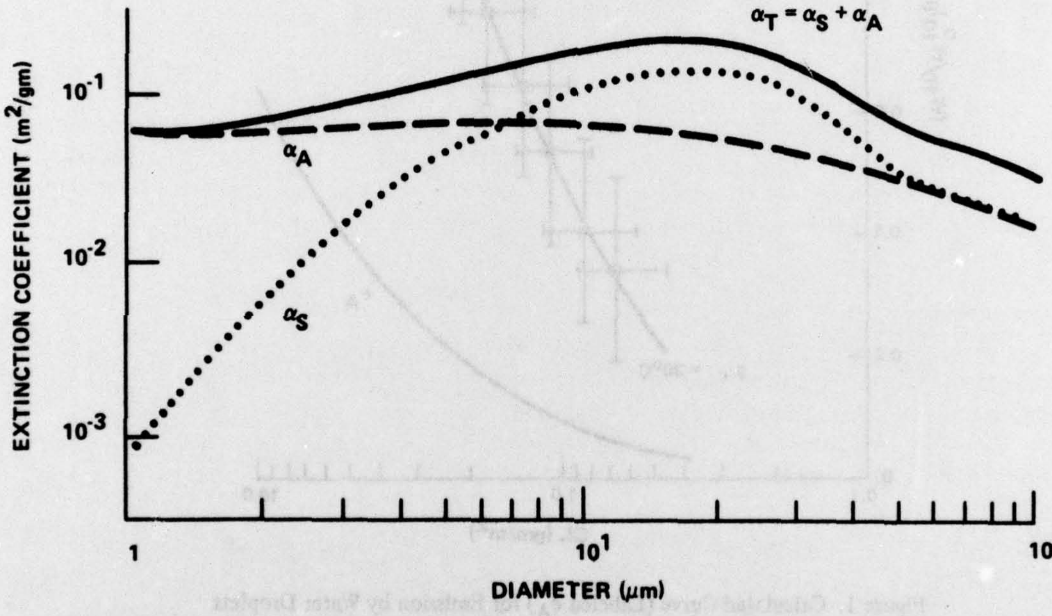

Figure 2. Computed Curves for Water Fog Showing Total Extinction Coefficient (Solid Curve) with Absorption (Dashed Curve) and Scattering (Dotted Curve) Contributions, as Functions of Droplet Diameter (Wavelength, λ, = 10 μm) (Reproduced from figure 9, reference 5).

From figure 2 at $\lambda = 10\ \mu m$, $\alpha_{A_{10}} \approx 0.06\ m^2/gm$ for 10-μm-diameter droplets, and equation 4 becomes

$$\epsilon_{10} = 1 - \exp(-\ 0.06\ CL) \tag{5}$$

Using equation 5, with a small additional correction for water vapor absorption at $\lambda = 10\ \mu m$, the curve labeled ϵ_A was computed for figure 1. The differences between emissances of the experimental (upper curve) and calculated (lower curve) values at various CL's in figure 1 thus represent emission due to water species other than water vapor or water droplets large enough to scatter visible light in cooling steam clouds. The magnitude of this difference is found to be dependent to some extent upon other environmental parameters. For example, an electrical

discharge in the chamber tends to shift the experimental curve upward, i.e., in the direction corresponding to greater activity of the "anomalous species."

In some experiments, the 30- to 40-minute cool-down period during which data such as those in figure 1 were taken was found to be abruptly terminated by a sudden loss of the anomalous emission component of the radiometric signal. Within 20 to 30 seconds, the emissance of these samples changed in a manner which could be represented by a point in figure 1 moving vertically downward from the lower end of the experimental curve to the equivalent abscissa value of the curve labeled ϵ_A, representing infrared emission by residual water droplets and vapor only. Apparently, this corresponded to a sudden reduction in the concentration of the "anomalous species" corresponding to the loss of saturation humidity.

Although most experimental data shown in figure 1 were taken for steam CL values of 0.6 to 2.0 gm/m^2 (C = 0.2 to 0.7 gm/m^3) corresponding to temperatures in the range $\theta_c = 30°$ to 39°C, the upper end of the experimental curve corresponds to extremely high CL values at temperatures near 100°C where, quite clearly, nonequilibrium conditions existed. In this region, radiant emittances up to about 1.4 times those of blackbodies at the same temperatures were found. Multiple optical scattering, which is not a factor at lower CL values, becomes significant under these conditions, and total emission simply exceeds that of an equivalent blackbody. This luminescence-like behavior cannot easily be related to the concentrations of steam producing it. Similar behavior of water near its boiling point was reported by others,[1] who attributed it to polymolecular water clusters of sizes 11 and 17.

IV. CONCLUSIONS.

Our results confirm the existence of strong infrared emission at the 10-μm wavelength from condensing and cooling steam clouds. This emission is anomalous in the sense that it cannot be accounted for by water vapor or droplets or by other commonly-known atmospheric constituents. Near 100°C and under nonequilibrium conditions, radiant emittance values exceeding those of blackbodies are observed, confirming observations by other workers of luminescence-like activity in water.

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GLOSSARY

A	Absorptance, the ratio of radiant energy absorbed by a body to that incident upon it, unitless.
α	Extinction or absorption coefficient (with subscript "A"), m^2/gm.
C	Aerosol concentration, gm/m^3.
D_μ	Geometric mean aerosol particle size diameter, μm.
ϵ	Emissance, ratio of the rate of radiant energy emission from a body at a given temperature to the corresponding rate emission from a blackbody at the same temperature, unitless.
L	Optical path length, m.
λ	Wavelength, μm.
Q	Mie efficiency factor, unitless.
ρ	Density of droplet liquid, gm/cm^3.
T	Transmittance, the ratio of radiant energy transmitted through a body or assemblage of bodies to that incident upon it/them, unitless.
θ_c	Temperature, °C.
W	Radiant emittance, watts/cm^2-μm. [Superscript "o" used with W denotes blackbody.]

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